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(54) Surface-roughened film and sheet, and process for production and use thereof.

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Description

The present invention relates to a novel surface-roughened film and sheet comprising a polymer of 4-methyl-1-pentene, and a process for the production and use thereof.

The essentially non-oriented surface-roughened film and sheet of the present invention is especially useful for the production of multilayer printed circuit boards, which are increasingly used in various kinds of electric apparatus. Usually, in a manufacturing process for multilayer printed circuit boards, first, single-sided metal-clad laminates or double-sided metal-clad laminates are produced and then an electric circuit is printed on those laminates, and the printed circuits are stacked via prepregs and pressed with heating to form the multilayer printed circuit boards. To strengthen the adhesion between the resin side of the single-sided metal-clad laminates and the prepregs, the surface of the resin side of the single-sided metal-clad laminate is preferably surface-roughened.

The above-mentioned single-sided or double-sided metal-clad laminate is manufactured by stacking a metal foil and a prepreg, and pressing them with heating to harden the prepreg. In such procedure, to surface-roughen the surface of the resin side of the laminate, a surface-roughened film or sheet is applied to a surface of the prepreg during the hardening process.

The surface-roughened film and sheet according to the present invention is useful as the above-mentioned surface-roughened film to roughen the surface of the resin side of the single-sided metal-clad laminate.

Japanese Examined Patent Publication No. 52-34664 discloses a process for the production of a biaxially oriented film having frosted surfaces comprising methyl-pentene polymer, which process comprises biaxially orienting a film comprising methylpentene polymer, high-density polyethylene and polystyrene, and heat-hardening the resulting film to form a frosted film. However the publication does not disclose the use of the film having frosted surfaces to surface-roughen the surface of the resin side of a laminate.

Japanese Unexamined Patent Publication No. 52-49823 discloses a process for the production of a laminate by an additive process characterized by placing a releasing film having a surface roughness of 0.5 to 5 µm on a prepreg impregnated with a thermoset resin, and forming it with heating. However the publication discloses only a triacetate film for the releasing film.

Japanese Unexamined Patent Publications No. 57-70653, No. 58-163648, and No. 57-70654 describe the use of 4-methylpentene-1 sheet as a releasing film in the manufacture of a laminate from a prepreg and metal foil. But these publications do not disclose the use of a surface-roughened film as a releasing film.

As a film providing roughness to the above-mentioned multilayer circuit board. TEDLAR (Trade mark of Du Pont, USA, a biaxially oriented film of polyvinyl fluoride containing a substantial amount of low molecular fillers and 5% of calcium carbonate is commercially available. Since, however, this film is produced by biaxial orientation, the surface thereof has deposits of the low molecular materials and calcium carbonate thereon. Therefore, when this film is used to produce single-sided metal clad-laminates by alternately stacking the copper foil and a prepreg, and the TEDLAR film between the layers of the copper foil and the prepreg, and pressing them with heating to harden the prepregs, the low molecular materials and the calcium carbonate may be transferred to the surface of the laminated copper foil, resulting in unsatisfactory adhesion between the copper surface and a resist, and unsatisfactory etching during further processing of the multilayer circuit boards.

Triacetylcellulose films having a surface roughened by sandblasting have been used. But, in this case, the fine sand and cellulose cause the same problems as mentioned above for TEDLAR.

The present invention provides an essentially non-oriented surface-roughened releasing film or sheet comprising a crystalline homopolymer or crystalline copolymer of 4-methyl-1-pentene having a mean surface roughness of 0.5 to 10 micrometers, wherein the polymer comprises at least 85 mol % 4-methyl-1-pentene units, and the surface-roughness is as produced by embossing a film or sheet of the polymer composition or by embossing a melt of the polymer composition; said film or sheet optionally containing up to 43 parts by weight of filler per hundred parts by weight of said polymer.

There is also provided a process for the production of the above-mentioned surface-roughened film or sheet comprising the following steps:

(1) preparing a molten composition comprising a homopolymer or copolymer of 4-methyl-1-pentene;

(2) extruding the molten composition to form a film or sheet; and

(3) placing the film or sheet in contact with a roll having a roughened surface or passing the film or sheet through a pair of rollers, at least one of the rollers having a roughened surface, wherein the mean surfac roughness of th surface of the roller is 0.5 to 10 micrometers, at a pressure and a temperature sufficient to replicate th roughness of the surface of the roll onto the surface of the film or sheet.

There is also provided a process for production of the above-mentioned surface roughened film or

sheet comprising the following steps:

(1) preparing a molten composition comprising a homopolymer or copolymer of 4-methyl-1-pentene;
(2) extruding the molten composition and
(3) applying the molten composition with a roll having a roughened surface or passing the molten composition through a pair of rollers, at least one of the rollers has a roughened surface, to form a film or sheet having a having a roughened surface, wherein the mean surface roughness of the roughened roller is 0.5 to 10 micrometers at a pressure and a temperature such that the roughness of the surface of the roll is replicated on the surface of the film or sheet.

Another object of the present invention relates to the use of the above-mentioned essentially non-oriented surface-roughened releasing film or sheet for the manufacture of single-sided metal clad laminates wherein the surface-roughened film or sheet is applied to a stack consisting of a prepreg and a metal foil so that the roughened surface of the surface-roughened film or sheet is in contact with a surface of the prepreg at a side opposite to the metal foil, and one or more of the stacks are placed between heat press plates and pressed with heating to harden the prepreg, wherein the roughness of the surface-roughened film or sheet is replicated onto the surface of the hardened prepregs.

BRIEF DESCRIPTION OF THE DRAWINGS

Figure 1 schematically represents a process for the production of a film or sheet of the present invention;
Fig. 2 schematically represents another process for the production of a film or sheet of the present invention;
Fig. 3 represents another system for the present process;
Figs. 4 and 5 represent processes for the production of single-sided metall-clad laminate the resin side of which is surface-roughened, through using a surface-roughened film of the present invention;
Fig. 6 represents the preparation of a test laminate; and
Fig. 7 represents general processes for the production of a multilayer printed circuit board.

BRIEF DESCRIPTION OF THE PREFERRED EMBODIMENTS

The film and sheet of the present invention comprises a crystalline homopolymer of 4-methyl-1-pentene, or a crystalline copolymer of 4-methyl-1-pentene with one or more of olefins. The polymer should have a high softening point so that it is fully resistant to a temperature at which a prepreg in question is hardened, and should be easily roughened at a temperature lower than a softening temperature of a roughening means. The olefins include α-olefin having 2 to 20 carbon atoms, preferably, ethylene, propylene, 1-butene, 1-hexene, 1-octene, 1-decene, 1-tetradecene, 1-hexadecene, and 1-octadecene. The poly4-methyl-1-pentene unit comprises at least 85 molar percent, preferably 90 to 98 molar percent of the copolymer. The melt flow rate of the poly4-methyl-1-pentene homopolymer or copolymer is generally 0.5 to 250 g/10 min., preferably 5 to 120 g/10 min., as measured under the condition of a load of 5 kg and a temperature of 260 °C. The melting viscosity of a polymer having a melt flow rate of less than 0.5 g/10 min. is too high, and results in a poor molding property; and the melting viscosity of a polymer having a melt flow rate of more than 200 g/10 min., is too low, and results in a low mechanical strength and poor molding properties.

The film and sheet may contain additives or fillers. One such additive is a silicone oil, which may improve the releasing property of the film from a hardened prepreg. The silicone oil has, for example, the following general formula:

$$R-\underset{R}{\overset{R}{\underset{|}{\overset{|}{Si}}}}-\left(O-\underset{R}{\overset{R}{\underset{|}{\overset{|}{Si}}}}-O\right)_{n}-\underset{R}{\overset{R}{\underset{|}{\overset{|}{Si}}}}-R$$

wherein R is the same or different, and represents a hydrogen atom, an aromatic hydrocarbon group, or a saturated or unsaturated aliphatic group optionally containing hetero atoms, and n represents a positive integer.

The aromatic hydrocarbon group includes an aromatic hydrocarbon group having 6 to 12 carbon atoms unsubstituted or substituted with lower alkyl groups having 1 to 3 carbon atoms, for example, phenyl, tolyl, thylphenyl, isopropylphenyl, naphthyl, and biphenyl.

The unsaturated or saturated aliphatic group includes, for example, alkyl groups having 1 to 6 carbon atoms, such as methyl, ethyl, propyl, butyl, pentyl, and hexyl; alkenyl or alkinyl groups having 2 to 6 carbon atoms, such as vinyl, 2-propenyl, propargyl, 3-butenyl, 4-pentenyl, and 5-hexenyl; and cycloaliphatic groups having 5 to 6 carbon atoms, such as cyclopentyl, cyclohexyl, cyclopentenyl, and cyclohexenyl.

The silicone oil preferably has a viscosity of 0•5 to 80,000 Pa.s, more preferably 5 to 50,000 Pa.s, as measured by rotational viscometer at 25°C.

The film or sheet contains the silicone oil preferably in an amount of 0.01 to 6% by weight, more preferably 0.5 to 5% by weight, in relation to the poly4-methyl-1-pentene polymer. The amount of the silicone oil should be limited as above mentioned so that the silicone oil is not transferred from the film or sheet to a surface of a hardened resin, resulting in a lowering of the adhesion strength between the hardened resin board of a single-sided metal-clad laminate and a prepreg when they are assembled.

Moreover, the film and sheet of the present invention can contain fillers, such as calcium carbonate, silica, mica, barium sulfate, talc and the like in an amount which does not lower the tear strength. Such fillers, if any, are used in an amount of not more than 43 parts by weight, preferably not more than 20 parts by weight, in relation to the weight of the poly4-methyl-1-pentene polymer. The fillers may provide an improved toughness to the film surface.

Finally, the film and sheet of the present invention may contain a thermostabilizing agent, weathering agent, rust preventive, copper inhibitor, antistatic agent, and the like, depending on the final use of the film or sheet.

The film and sheet of the present invention have a roughened-surface on one or both sides thereof. The roughness is transferred or replicated, for example, to a surface of a resin side of a single-sided metal-clad laminate. The mean surface roughness is 0.5 to 10 micrometers. A mean surface roughness of less than 0.5 micrometers cannot provide a sufficient surface area for improving the adhesion between the hardened resin and a prepreg. On the other hand, a mean surface roughness of more than 10 micrometers will provide a too strong adhesion between the hardened resin and the film of the present invention, resulting in breakage of or partial adhesion to the hardened resin of the film when it is released. The maximum surface roughness is preferably less than 20% of the thickness of the film or sheet.

According to the present invention, the mean surface roughness is represented by a center line mean roughness (Ra) defined by JIS B0601-1970, and is measured, for example, by a Surface Figure Measuring Apparatus (Type SE-3A) provided by K.K. Kosaka Kenkyusho in Japan.

The thickness of the film or sheet of the present invention is usually 5 to 1000 micrometers, and preferably 15 to 200 micrometers.

The film and sheet of the present invention are produced from a composition comprising a poly4-methyl-1-pentene homo- or copolymer, and optionally, additives and/or fillers.

The composition is molten, for example at 270°C to 320°C, and extruded from a die.

After that, a surface-roughened film or sheet is produced according to one of two procedures.

According to one procedure, the molten composition is applied to a roll having a roughened surface, so that the film or sheet is formed and simultaneously the film or sheet is surface-roughened. The procedure is schematically shown in Figs. 1 and 2.

According to another procedure, the composition is extruded to form a film or sheet, and then the film or sheet is surface-roughened. This procedure is schematically shown in Fig. 3.

Referring to Fig. 1, the apparatus used for production of the present film or sheet comprises a T-die 1 and a pair of rolls 2a and 2b adjacent to an extruder outlet of the T-die. At least one of the rolls has a roughened surface 4a with a mean surface roughness of 0.5 to 10 micrometers.

A molten composition is extruded through the T-die in a conventional fashion, and is passed between the rolls having, for example, 40 to 100°C of temperature at a pressure of 4 to 41 bar (3 to 40 kg/cm^2) to form a film or sheet having a roughened surface and the roughened film or sheet 5 is wound by wind-up roll (not shown). Where both the rolls have a roughened surface, a film or sheet roughened on both sides is produced.

Referring to Fig. 2, the apparatus comprises a T-die 1, a roll 2c having a roughened film 4b adhered thereon, and a roll 6. The roughened film 4b has surface roughness of 0.5 to 10 micrometers, and is made from a material having a softening temp rature higher than that of the film or sheet 5 of the present inv ntion. The mat rial for th roughened film 4b includes, for example, fluorine plastics such as ethylene tetrafluoride resin, polyvinylid ne fluorid etc, polysulfone, and polyvinyl ether ketone, and the like.

Referring to Fig. 3, a combination of a haul-off apparatus 3 and a surface treatment apparatus 7 is used. As seen from Fig. 3, the molten composition is extruded through the T-die 1, the formed film or sheet is cooled in the haul-off apparatus 3, and the film or sheet is then sent from the haul-off apparatus 3 to the surface treatment apparatus 7. The surface treatment apparatus 7 contains heating rolls 8 and a pair of

surface roughening rolls 2d and 2e, at least one of which has a roughened surface with a roughness of 0.5 to 10 micrometers. The cooled film or sheet provided from the haul-off apparatus 3 is again heated by the heating rolls 8 to a temperature at which th roughness of the surface roughening rolls 2d and 2e can be replicated onto the heated film or sheet, i.e., to a temperature higher than the softening temperature of the film or sheet, for example 140 to 220°C. Finally the heated film or sheet is passed between the paired roughening rolls 2d and 2e at a pressure of 41-101 bar (40 to 100 kg/cm²) to provide the roughness to the film or sheet. If only one of the roughening rolls 2d and 2e has the roughened surface, a film or sheet roughened on only one side is obtained; and if both roughening rolls 2d and 2e have a roughened surface, a film or sheet roughened on both sides is obtained. Alternatively, the surface treating apparatus 7 can contain one roughening roll as shown in Fig. 2 rather than a pair of the roughening rolls.

The roughening roll per se may have a roughened surface as shown in Fig. 1. Alternatively, the roughening roll comprises a roll without a surface roughness and a roughened film is adhered to the surface of the roll, as shown in Fig. 2. In the latter case, the present process can be economically carried out using any conventional film or sheet forming apparatus, by modifying a roll by adhering a surface roughened film thereon.

The film and sheet of the present invention is especially useful in the manufacture of multilayer printed circuit boards.

Figure 7 explains a general process for the manufacture of multilayer printed circuit boards. One or more internal layer circuit boards 23 or 33, prepregs 22 or 32, a pair of single-sided or double-sided metal clad laminates 21a and 21b, or 31a and 31b and outside tools 24 or 34 are stacked as shown in Fig. 7(a) or 7(b), the stack is placed between two heating press plates 26 or 36 via cushion materials 25 or 35, and the stack is pressed with heating to harden the prepregs, resulting in a firmly integrated laminate which is then processed by perforation, through-hole plating, and surface etching to complete a multilayer printed circuit board.

The adhesion between the prepreg 22 and the surfaces of the resin sides of the above-mentioned single-sided metal-clad laminates or internal layer circuit boards 21a, 21b and 23 is improved by providing a roughness on the surface of the resin board.

Note, the above-mentioned single-sided or double-sided metal laminates 21a, 21b, 31a and 31b, and the internal layer circuit boards 23 and 33 are manufactured by stacking a copper foil and prepreg, and pressing the stack with heating to harden the prepreg, resulting in adhesion between the copper foil and the resin board of the hardened prepreg. In such a process, by applying a surface roughened film or sheet to the surface of the prepreg at the side opposite to the copper foil, the single-sided copper-clad laminated having the surface roughened resin side can be obtained. The film or sheet of the present invention is preferably as the surface roughened film used in this process.

In many cases, more than one of the above-mentioned stacks consisting of the copper foil and the prepreg are stacked, and pressed with heating to manufacture more than one metal-clad laminate simultaneously. In such a case, the surface roughened film also serves as a releasing material.

Examples

The present invention will now be further illustrated by, but is by no means limited to, the following examples.

Example 1

Referring to Fig. 3, a crystalline copolymer of 4-methyl-1-pentene, 1-hexadecene and 1-octadecene, having 95 mole percents of 4-methyl-1-pentene unit, and a melt flow rate of 26 g/10 min., was melted with a small amount of phenolic antioxidant in an extruder 9 having a diameter of 65 mm at 280°C, the molten polymer was extruded through a manifold T-die 1, and the extruded polymer was cooled by cooling rolls at 60°C to form a film having a thickness of 50 micrometers.

The film thus prepared was then heated by heating rolls 8 at 200°C, and the heated film was passed through a pair of roughening rolls 2d and 2e to roughen the surface of the film.

As shown in Fig. 4, the surface-roughened film 13a thus prepared, glass-reinforced epoxy resin prepregs having a thickness of 500 micrometers 12a and 12b, and copper foils 11a and 11b having a thickness of 40 micrometers w re stacked one upon th oth r. Tools 15 were applied to both sides of the stack, and the stack placed between two heating press plates 17 via cushion materials 16. The stack was preheated by the press plates 17 at 180°C, at a pressure of 6 bar (5 kg/cm²·G) for 3 minutes, and then pressed at a pressure of 31 bar (30 kg/cm²·G) for another 3 minutes to harden the epoxy prepreg, and thus

two single-sided metal-clad laminates were produced.

Next, as shown in Fig. 5, the single-sided metal-clad laminates 11a/12a and 11b/12b were separated from the surface roughened film 13a. As shown in Fig. 6, the single-sided metal-clad laminates 11a/12a and 11b/12b were stacked via a prepreg 12c, and the stack was placed between heating press plates via tools 15 and cushions 16, and the prepreg was hardened as described above to form a multilayer board.

Note that although the above-mentioned examples represent a model for the manufacture of a multilayer board, in practice, in the manufacture of a multilayer printed circuit board as shown in Fig. 7a and 7b, one or more internal layer circuit boards are stacked between the single-sided metal-clad laminates.

The single-sided metal-clad laminate and the multilayer board prepared as above were evaluated as follows:

The peel strength N/1.5 mm (kg/15 mm) between the single-sided metal-clad laminate and the surface roughened film, and the peel strength N/1.5 mm (kg/15 mm) between the hardened epoxy resin 12a of the single-sided metal-clad laminate and the hardened resin 12c converted from the epoxy prepreg were measured by using an peeling test equipment (Instron Type Universal Tester made by Instron Company in U.S.A.) under a peeling speed of 200 mm/min.

The physical properties were evaluated as follows:

(1) Gloss: ASTM-D2457 (angle 60°)

(2) Surface roughness: Surface Figure Measuring Apparatus (K.K. Kosaka Kenkyusho)

(3) Tear Strength: ASTM-D1922

The results are set forth in Table.

Example 2

The procedure of Example 1 was repeated except that a roll with the surface roughness of 2 micrometers was used. The results are set forth in the Table.

Example 3

The procedure of Example 1 was repeated except that a roll having a surface roughness of 7 micrometers was used. The results are set forth in the Table.

Example 4

The procedure of the Example 1 was repeated except that the roughening roll was prepared by adhering a surface roughening film to the roll as shown in Fig. 2. The results are set forth in the Table.

Example 5

The procedure of Example 1 was repeated except that 5.3 parts by weight of calcium carbonate was mixed with 100 parts by weight of the copolymer of 4-methyl-1-pentene. The results are set forth in the Table.

Comparative Example 1 ("Co-Ex 1")

A film extruded from the T-die under the condition described in Example 1, but not surface-roughened, was used to prepare a multilayer printed circuit board. The results are set forth in the Table.

Comparative Example 2 ("Co-Ex 2")

The procedure of Example 1 was repeated except that a surface-roughened film having a mean roughness of 20 micrometers was used for the roughening roll. The results are set forth in the Table.

Comparative Exampl 3 ("Co-Ex 3")

The procedure of Example 1 was repeated except that 66.7 parts by weight of calcium carbonate was mixed with 100 parts by weight of the copolymer of 4-m thyl-1-pentene. The results are set forth in the Table.

Table

		Ex. 1	Ex. 2	Ex. 3	Ex. 4	Ex. 5	Co-Ex. 1	Co-Ex. 2	Co-Ex. 3
Gloss (%)	Surface-roughened film	6	10	4	7	5	95	24	2
	Hardened prepreg	7	13	5	8	6	90	27	3
Surface roughness (micro meter)	Mean roughness	6	3	8	5	7	0.1	25	8
	Difference between max. and min.	10	5	12	8	10	0.3	50	10
Peel strength (1) N/1.5 mm (kg/15 mm)		0.05	0.03	0.06	0.04	0.1	0.01	1.1	1.2
Elmendorf tear strength N/mm (kg/cm)	MD	35	35	35	35	30	35	35	3
	TD	80	80	80	80	75	80	80	5
Peel strength (2) N/1.5 mm (kg/15 mm)		3.5	2.8	4.0	3.1	3.8	0.8	5.5	4.0

Peel strength (1) : between a single-sided metal-clad laminate (copper foil/hardened epoxy resin) and a surface-roughened film.

Peel strength (2) : between a hardened epoxy resin layer of a single-sided metal-clad laminate and a prepreg.

As seen from th Table, in Examples 1 to 5, the peel strength between the hardened epoxy resin layer and the surface-roughened film is as low as 0.03 to 0.1 N/1.5 mm (kg/15 mm), thus showing an improved rel ase property. Also, the peel strength between the surface-roughened poxy resin layer and the hard ned pr preg is as high as 2.8 to 4.0 N/1.5 mm (kg/15 mm), showing an improv d adherence.

On th other hand, the film having a smooth surface provides a better release property but does not improve th adhesion between th hardened resin layer and the prepreg. Th adhesion is as low as 0.8

N/1.5 mm (kg/15 mm), as seen from Comparative Example 1.

Moreover, as seen from Comparative Example 2, the film having a surface roughness as high as 25 micrometers provides a poor release property as represented by a high peel strength of 1.1 N/1.5 mm (kg/15 mm); and as seen from Comparative Example 3, a film containing a too many amount of filler provides a poor release property, as represented by a high peel strength of 1.2 N/1.5 mm (kg/15 mm) and a low tear strength of 3 to 5 N/mm (kg/cm).

Since the film and sheet of the present invention comprises a crystalline homopolymer or crystalline copolymer of 4-methyl-1-pentene, it exhibits preferable properties inherent in a polymer comprising 4-methyl-1-pentene, i.e., excellent hardness, impact resistance, and heat resistance. Moreover, since the film or sheet of the present invention do not contain a substantial amount of inorganic fillers, they are not easily torn or broken, and do not deposit impurities on their surface.

Therefore, when used for the manufacture of a single-sided metal-clad laminate as a surface-roughened film or sheet, they do not soften and deteriorate during the hardening of the prepreg, and are easily peeled from the hardened resin. Since they do not deposit impurities thereon, therefore the surface of copper foil is free from impurities which would interfere with the etching process.

Moreover, since the film and sheet of the present invention have at least one roughened surface having a surface roughness of 0.5 to 10 micrometers, an improved release property is provided between the hardened and surface-roughened resin and the film or sheet, and improved adhesion is provided between the hardened and surface-roughened resin of the laminate and a hardened prepreg.

Therefore the present film and sheet are especially useful in the manufacture of metal-clad laminates, and therefore, of multilayer printed circuit boards.

Claims

1. An essentially non-oriented surface-roughened releasing film or sheet comprising a crystalline homopolymer or crystalline copolymer of 4-methyl-1-pentene having a mean surface roughness of 0.5 to 10 micrometers, wherein the polymer comprises at least 85 mol % 4-methyl-1-pentene units, and the surface-roughness is as produced by embossing a film or sheet of the polymer composition or by embossing a melt of the polymer composition; said film or sheet optionally containing up to 43 parts by weight of filler per 100 parts by weight of said polymer.

2. A surface-roughened film or sheet according to claim 1 wherein the copolymer consists of 4-methyl-1-pentene and an α-olefin selected from the group consisting of ethylene, propylene, 1-butene, 1-hexene, 1-octene, 1-decene, 1-tetradecene, and 1-octadecene.

3. A surface-roughened film or sheet according to claim 1 or 2 wherein the film or sheet further contains 0.5 to 5% by weight of silicone oil in relation to the weight of the polymer of 4-methyl-1-pentene.

4. A surface-roughened film or sheet according to any preceding claim for use in the manufacture of single-sided metal-clad laminates as a roughening means.

5. A process for the production of an essentially non-oriented surface roughened releasing film or sheet comprising a crystalline homopolymer or crystalline copolymer of 4-methyl-1-pentene having a mean surface roughness of 0.5 to 10 micrometers wherein the polymer comprises at least 85 mol % 4-methyl-1-pentene units, comprising the steps of:
(1) preparing a molten composition comprising a homopolymer or copolymer of 4-methyl-1-pentene; (2) extruding the molten composition to form a film or sheet; and (3) placing the film or sheet in contact with a roll having a roughened surface or passing the film or sheet through a pair of rollers, at least one of the rollers has a roughened surface wherein the mean surface roughness of the roughened roller is 0.5 to 10 micrometers, at a pressure and a temperature such that the roughness of the surface of the roll is replicated on the surface of the film or sheet.

6. A process for production according to claim 5 wherein the surface-roughened roller comprises a roller and a surface-roughened film or sheet adhered to the surface of the roller.

7. A process for the production of an essentially non-oriented surface-roughened releasing film or sheet comprising a crystalline homopolymer or crystalline copolymer of 4-methyl-1-pentene having a mean surface roughness of 0.5 to 10 micrometers wherein the polymer comprises at least 85 mol % 4-

methyl-1-pentene units, comprising the steps of:

(1) preparing a molten composition comprising a homopolymer or copolymer of 4-methyl-1-pentene;
(2) extruding the molten composition; and
(3) applying the molten composition with a roll having a roughened surface or passing the molten composition though a pair of rollers, at least one of which rollers has a roughened surface, to form a film or sheet having a roughened surface, wherein the mean surface roughress of the roughened roller is 0.5 to 10 micrometers at pressure and a temperature such that the roughness of the surface of the roll is replicated on the surface of the film or sheet.

8. A process for production according to claim 7 wherein the surface-roughened roller comprises a roller and a surface-roughened film or sheet adhered to the surface of the roller.

9. Use of an essentially non-oriented surface-roughened releasing film or sheet of claim 1 for manufacture of single-sided metal clad laminates wherein the surface-roughened film or sheet is applied to a stack consisting of a prepreg and a metal foil so that the roughened surface of the surface-roughened film or sheet is in contact with a surface of the side of the prepreg opposite to the metal foil, and one or more of the stacked laminates are placed between heat press plates and pressed with heating to harden the prepregs, wherein the roughness of the surface-roughened film or sheet is replicated on the surface of the hardened prepregs.

Revendications

1. Un film ou une feuille antiadhérent à surface rendue rugueuse essentiellement non orientée comprenant un homopolymère cristallin ou un copolymère cristallin du 4-méthyl-1-pentène ayant une rugosité moyenne de surface de 0,5 à 10 µm, selon lequel le polymère comprend au moins 85 % molaires d'unités 4-méthyl-1-pentène, et la rugosité de surface est produite par gaufrage d'un film ou d'une feuille de la composition de polymère ou par gaufrage d'une masse fondue de la composition de polymère, ledit film ou feuille contenant éventuellement jusqu'à 43 parties en poids de charges pour 100 parties en poids dudit polymère.

2. Un film ou une feuille à surface rendue rugueuse selon la revendication 1, selon lequel le copolymère est constitué de 4-méthyl-1-pentène et d'une α-oléfine choisie dans le groupe comprenant l'éthylène, le propylène, le 1-butène, le 1-hexène, le 1-octène, le 1-décène, le 1-tétradécène et le 1-octadécène.

3. Un film ou une feuille à surface rendue rugueuse selon la revendication 1 ou 2, selon lequel le film ou la feuille contient en outre 0,5 à 5 % en poids d'huile de silicone par rapport au poids du polymère de 4-méthyl-1-pentène.

4. Un film ou une feuille à surface rendue rugueuse conformément à l'une quelconque des revendications précédentes à utiliser dans la fabrication des stratifiés métallisés sur un seul côté en tant que moyen pour former des rugosités.

5. Un procédé pour produire un film ou une feuille antiadhérent à surface rendue rugueuse essentiellement non orientée comprenant un homopolymère cristallin ou un copolymère cristallin du 4-méthyl-1-pentène ayant une rugosité moyenne de surface de 0,5 à 10 µm, selon lequel le polymère comprend au moins 85 % molaires d'unités 4-méthyl-1-pentène, comprenant les étapes suivantes :

(1) préparation d'une composition fondue comprenant un homopolymère ou un copolymère du 4-méthyl-1-pentène ; (2) extrusion de la composition fondue pour former un film ou une feuille ; et (3) placement du film ou de la feuille en contact avec un cylindre ayant une surface rugueuse ou passage du film ou de la feuille entre une paire de cylindres, dont l'un au moins a une surface rugueuse, selon lequel la rugosité moyenne de surface du cylindre rugueux est de 0,5 à 10 µm, à une pression et à une température telles que la rugosité de la surface du cylindre soit reproduite sur la surface du film ou de la feuille.

6. Un procédé de production selon la revendication 5, selon lequel le cylindre à surface rugueuse comprend un cylindre et un film ou une feuille à surface rugueuse adhérent à la surface du cylindre.

7. Un procédé de production d'un film ou d'une feuille antiadhérent à surface rendu rugueuse essentiel-

lement non orientée comprenant un homopolymère cristallin ou un copolymère cristallin du 4-méthyl-1-pentène ayant une rugosité moyenne de surface de 0,5 à 10 µm selon lequel le polymère comprend au moins 85 % molaires d'unités 4-méthyl-1-pentène, comprenant les étapes suivantes :

(1) préparation d'une composition fondue comprenant un homopolymère ou un copolymère du 4-méthyl-1-pentène ; (2) extrusion de la composition fondue ; et (3) application à la composition fondue d'un cylindre ayant une surface rugueuse ou passage de la composition fondue entre une paire de cylindres, dont l'un au moins a une surface rugueuse, pour former un film ou une feuille ayant une surface rendue rugueuse, selon laquelle la rugosité moyenne de surface du cylindre rugueux est de 0,5 à 10 µm à une pression et à une température telles que la rugosité de la surface du cylindre soit reproduite sur la surface du film ou de la feuille.

8. Un procédé de production selon la revendication 7, selon lequel le cylindre à surface rugueux comprend un cylindre et un film ou une feuille à surface rugueuse adhérent à la surface du cylindre.

9. Utilisation d'un film ou d'une feuille antiadhérent à surface rendue rugueuse essentiellement non orientée selon la revendication 1 pour la fabrication de stratifiés métallisés sur un seul côté selon laquelle le film ou la feuille à surface rendue rugueuse est appliqué à une pile constituée d'un préimprégné et d'une feuille métallique de sorte que la surface rugueuse du film ou de la feuille à surface rendue rugueuse est en contact avec une surface du côté du préimprégné opposé à la feuille métallique, et un ou plusieurs stratifiés empilés sont placés entre les plateaux de presse chauffante et pressés avec chauffage pour durcir les préimprégnés, selon laquelle la rugosité du film ou de la feuille à surface rendue rugueuse est reproduite sur la surface des préimprégnés durcis.

Patentansprüche

1. Ein im wesentlichen nicht gerichteter, eine rauhe Oberfläche besitzender Ablösefilm oder ein im wesentlichen nicht gerichtetes, eine rauhe Oberfläche besitzendes Ablöseblatt mit einem kristallinen Homopolymer oder einem kristallinen Copolymer von 4-Methyl-1-penten und einer mittleren Oberflächen-Rauhigkeit von 0,5 bis 10 Micrometern, bei dem das Polymer mindestens 85 Mol % 4-Methyl-1-penten-Einheiten aufweist und die Oberflächen-Rauhigkeit durch Prägen eines Films oder Blattes der Polymer-Zusammensetzung oder durch Prägen einer Schmelze der Polymer-Zusammensetzung erzeugt wird; wobei der Film oder das Blatt wahlweise bis zu 43 Gewichtsteile eines Füllstoffs pro 100 Gewichtsteile des Polymers enthält.

2. Ein eine rauhe Oberfläche besitzender Film oder Blatt nach Anspruch 1, bei dem das Copolymer besteht aus 4-Methyl-1-penten und einem α-Olefin, das ausgewählt ist aus der Gruppe, die besteht aus Ethylen, Propylen, 1-Buten, 1-Hexen, 1-Octen, 1-Decen, 1-Tetradecen und 1-Octadecen.

3. Ein eine rauhe Oberfläche besitzender Film oder Blatt nach Anspruch 1 oder 2, bei dem der Film oder das Blatt weiterhin 0,5 bis 5 Gewichtsprozent Siliconöl bezogen auf das Gewicht des 4-Methyl-1-penten-Polymers enthält.

4. Ein eine rauhe Oberfläche besitzender Film oder Blatt nach irgendeinem der vorhergehenden Ansprüche zum Gebrauch als eine Rauhigkeit schaffende Einrichtung bei der Herstellung von einseitig metallplattierten Laminaten.

5. Ein Verfahren zur Herstellung eines im wesentlichen nicht gerichteten, eine rauhe Oberfläche besitzenden Ablöse-Films oder Blattes mit einem kristallinen Homopolymer oder einem kristallinen Copolymer von 4-Methyl-1-penten und einer mittleren Oberflächen-Rauhigkeit von 0,5 bis 10 Micrometern, bei dem das Polymer mindestens 85 Mol % 4-Methyl-1-penten-Einheiten aufweist, das folgende Schritte aufweist:

(1) Herstellen einer geschmolzenen Zusammensetzung mit einem Homopolymer oder einem Copolymer von 4-Methyl-1-penten;
(2) Pressen der geschmolzenen Zusammensetzung zur Bildung eines Films oder eines Blattes; und
(3) Anordnen des Films oder Blatts in Kontakt zu einer Walze mit rauher Oberfläche oder Durchlassen des Films oder des Blatts durch ein Walzenpaar, wobei mindestens eine der Walzen eine rauhe Oberfläche besitzt und die mittlere Oberflächen-Rauhigkeit der rauhen Walze 0,5 bis 10

Micrometer ist, bei einem derartigen Druck und einer derartigen Temperatur, daß die Rauhigkeit der Walzenoberfläche auf der Oberfläche des Films oder des Blattes wiedergegeben wird.

6. Ein Herstellungsverfahren nach Anspruch 5, bei dem die Walze mit rauher Oberfläche eine Walze und einen an der Oberfläche der Walze anhaftenden Film oder ein an der Oberfläche der Walze anhaftendes Blatt mit rauher Oberfläche umfaßt.

7. Verfahren zur Herstellung eines im wesentlichen nicht gerichteten, eine rauhe Oberfläche besitzenden Ablöse-Films oder Blattes mit einem kristallinen Homopolymer oder einem kristallinien Copolymer von 4-Methyl-1-penten und einer mittleren Oberflächen-Rauhigkeit von 0,5 bis 10 Micrometern, bei dem das Polymer mindestens 85 Mol % 4-Methyl-1-penten-Einheiten aufweist, das folgende Schritte aufweist:

   (1) Herstellen einer geschmolzenen Zusammensetzung mit einem Homopolymer oder Copolymer von 4-Methyl-1-penten;
   (2) Fressen der geschmolzenen Zusammensetzung; und
   (3) Inkontaktbringen der geschmolzenen Zusammensetzung mit einer eine rauhe Oberfläche besitzenden Walze oder Durchlassen der geschmolzenen Zusammensetzung durch ein Walzenpaar, wobei mindestens eine der Walzen eine rauhe Oberfläche besitzt, um einen Film oder ein Blatt mit einer rauhen Oberfläche zu bilden, wobei die mittlere Oberflächen-Rauhigkeit der rauhen Walze 0,5 bis 10 Micrometer ist, bei einem solchen Druck und einer solchen Temperatur, daß die Rauhigkeit der Walzen-Oberfläche auf der Oberfläche des Films oder des Blattes wiedergegeben wird.

8. Herstellungsverfahren nach Anspruch 7, bei dem die Walze mit rauher Oberfläche eine Walze und einen an der Oberfläche der Walze anhaftenden Film oder ein an der Oberfläche der Walze anhaftendes Blatt mit rauher Oberfläche umfaßt.

9. Verwendung eines im wesentlichen nicht gerichteten, eine rauhe Oberfläche besitzenden Ablöse-Films oder Blattes nach Anspruch 1 zur Herstellung von einseitig metallplattierten Laminaten, bei der der Film oder das Blatt mit rauher Oberfläche dergestalt in einer Stapelung aus einem Prepreg und einer Metallfolie angebracht wird, daß die rauhe Oberfläche des Films oder Blatts mit rauher Oberfläche in Kontakt ist mit einer Oberfläche der von der Metallfolie abgewandten Seite des Prepreg, und bei der ein oder mehrere gestapelte Laminate zwischen den Platten einer Wärmepresse angeordnet werden und unter Erhitzen gepreßt werden, um die Prepregs zu härten, wobei die Rauhigkeit des Films oder Blattes mit rauher Oberfläche auf der Oberfläche der gehärteten Prepregs wiedergegeben wird.

Fig. 1

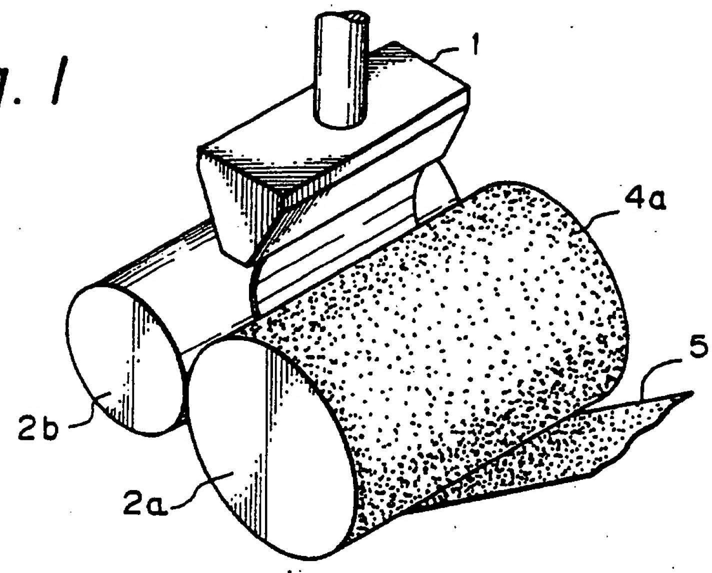
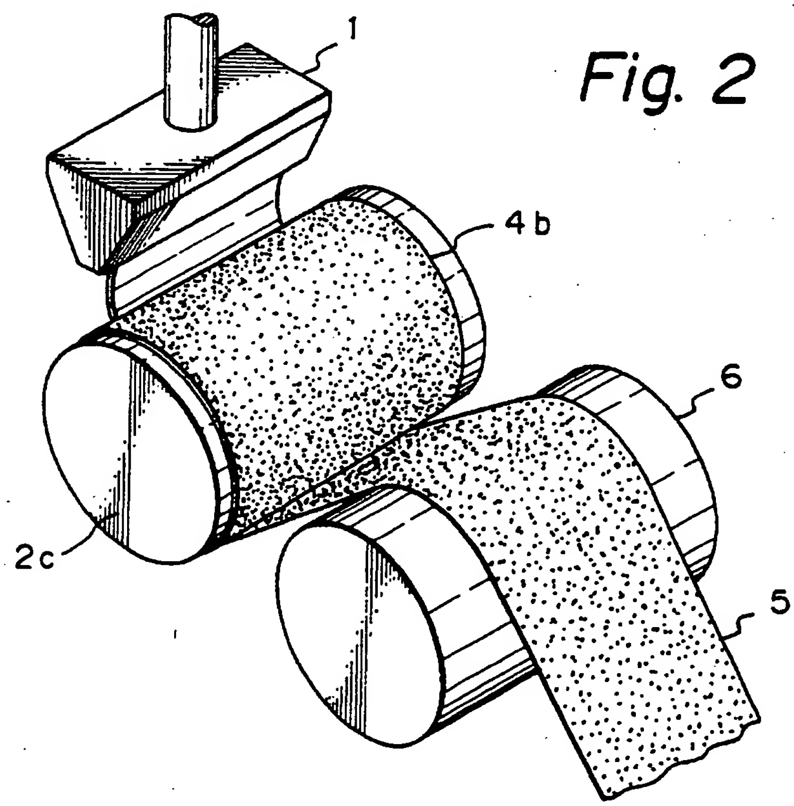


Fig. 2



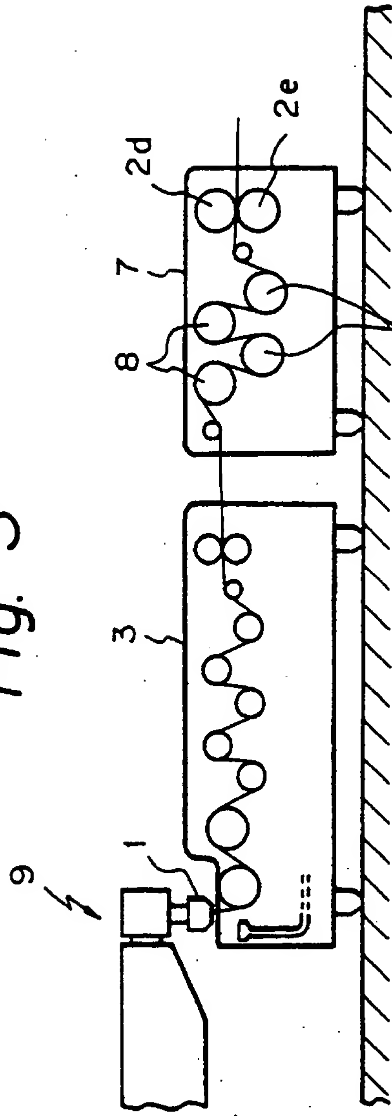


Fig. 3

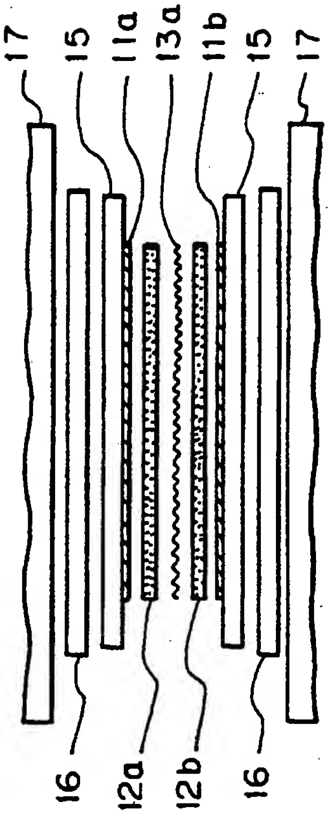


Fig. 4

Fig. 5

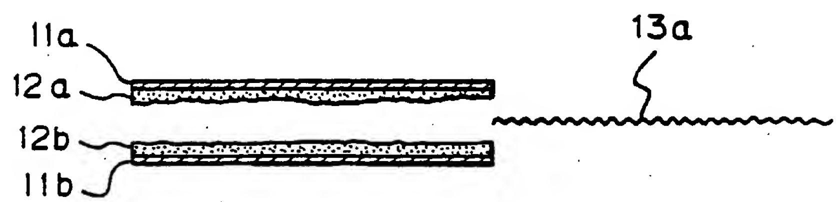


Fig. 6

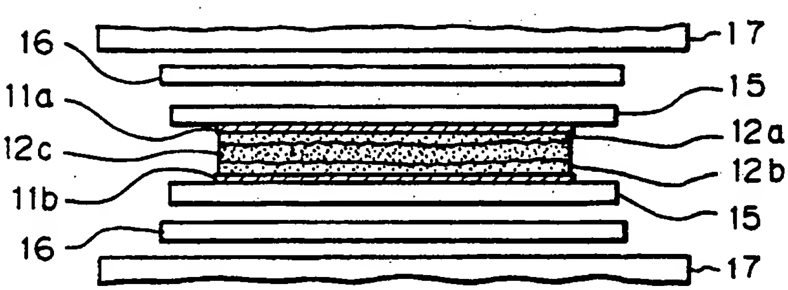


Fig. 7b

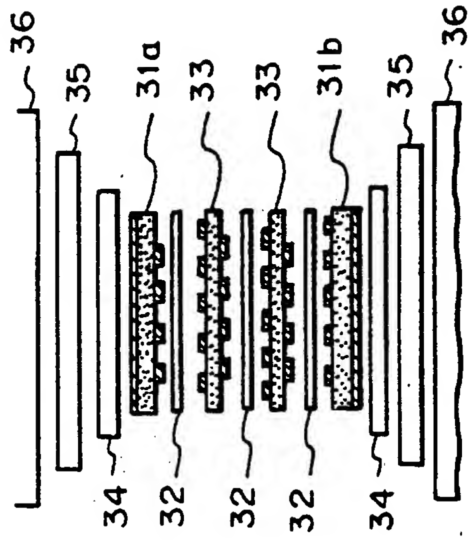

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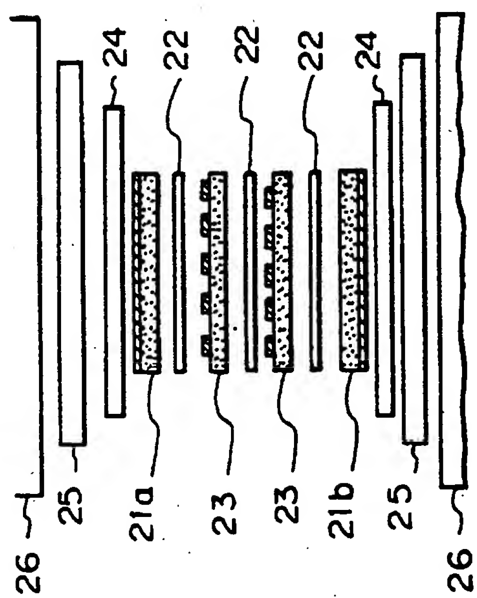
Fig. 7a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